Charge-transfer model of *s*- and *d*-wave pairing in the cuprates

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Local carrier pairs in CuO_2 planes are coupled to neutralizing displacement charges described by doubly charged local bosons which are identified with a feature seen in optical experiments while square symmetry determines the pair wave function to be a superposition of *s* and *d* waves. Within this model transition temperatures T_{cs} and T_{cd} are determined in weak coupling and a general form of the Ginzburg-Landau free energy is derived. Finally a strong-coupling formalism is set up which sheds new light on the gap equations and determines the properties of the local boson. [S0163-1829(96)01830-9]

I. INTRODUCTION

The pairing symmetry in the cuprate superconductors is a controversial issue of present day research. Indeed, while many experiments are in accord with conventional *s*-wave pairing there is an important body of data which seems to be compatible only with *d*-wave symmetry.[1,2] Because of this situation the idea of the simultaneous presence of both, *s*- and *d*-wave order parameters but a unique transition temperature seems quite natural.[3] This situation renders the already considerable complexity of the cuprates and of their properties even more serious so that the hope of an explanation by one single model seems rather unrealistic. Instead, a phenomenological description in terms of a Ginzburg-Landau free-energy functional generalized so as to incorporate both, *s*- and *d*-wave order parameters[4] but supplemented by minimal models defining the physical parameters seems to be most appropriate. In this spirit and also in order to render the theoretical steps more transparent we here renounce numerical computations in favor of approximate analytical calculations.

The short coherence lengths and the proximity of a Mott transition but also optical experiments suggest that a tight-binding approximation is an appropriate description of the superconductivity in the cuprates. In particular, the coupling between carriers seems to be well described by a charge-transfer mechanism.[5,6] Here we apply this point of view to the picture of hole pairs in real space (CuO_2 planes) described by a pair wave function of the size of the coherence length, as introduced in the earlier discussion of the cross-over problem between weak and strong coupling.[7,8] Retaining only nearest-neighbor pairings and imposing square symmetry, an explicit form of the pair wave function is obtained which automatically incorporates also short-range antiferromagnetic spin correlations as evidenced in nuclear magnetic resonance.[9] These local carrier pairs are then coupled to neutralizing polarization charges described by a doubly charged, local boson field (Sec. II). It turns out that this coupling is a generalization to the case of combined *s*- and *d*-wave symmetry of an earlier model by the author.[10,11] Elimination of the charged local boson field gives rise to an effective carrier attraction[11] which, in weak-coupling approximation, leads to gap equations for *s*- and *d*-symmetry and their combination. Using as a cutoff the experimental value for the boson linewidth from Ref. 6, transition temperatures T_{cs} and T_{cd} are then calculated (Sec. III). This local approach has similarities to the work of Sörensen and co-workers.[12]

In a further step the effective attraction is eliminated by a Hubbard-Stratonovich transformation[13,14] in order to obtain an effective action as function of the two order parameters with *s* and *d* symmetry. Evaluation of this action to fourth order in the order parameters then yields a time-dependent Ginzburg-Landau functional. And restricting this functional to equilibrium and imposing square symmetry we obtain an expression for the Ginzburg-Landau free energy which has the same generality as the phenomenological form derived by Sigrist and co-workers[4] (Sec. IV). Thus our results may directly be used for the interpretation of experimental data thereby yielding information about the parameters of the model.

In a final step the self-energies of the carriers and of the local charged boson are calculated, the former being used in a strong-coupling derivation of the gap equations,[11] while the latter serves to determine the properties of the boson (Sec. V). Finally, it is argued (Sec. VI) that the simple model presented in this paper exhibits many of the essential features a theory of superconductivity in the cuprates must possess and that generalizations by inclusion of the third dimension in the spirit of Anderson and co-workers[15,12] and of more than nearest-neighbor pairings[12] are fairly straightforward. Since charge couplings involve much higher energies than spin couplings it is argued that the present theory may be considered a serious alternative to models in which pairing is mediated by antiferromagnetic spin fluctuations.[16]

II. CHARGE-TRANSFER HAMILTONIAN

The most general singlet pair operator has the form[7,8,14]

$$B_{\mathbf{q}}^{+}=\sum_{\mathbf{k}}\ \varphi_{\mathbf{k}-\mathbf{q}/2}a_{\mathbf{k}\uparrow}^{+}a_{\mathbf{q}-\mathbf{k}\downarrow}^{+}\,, \tag{2.1}$$

where $\tilde{\varphi}(\mathbf{r})=V^{-1/2}\Sigma_{\mathbf{k}}\varphi_{\mathbf{k}}e^{i\mathbf{k}\cdot\mathbf{r}}$ is the pair wave function, $V=La^2$ is the (two-dimensional) crystal volume, and a is the lattice constant of the (quadratic) CuO_2 lattice. (We do not consider orthorhombic distortions[4] in this paper.) The coupling between the carrier pairs and the doubly charged local boson field then may be written as

$$H' = \gamma \sum_{\mathbf{q}} B_{\mathbf{q}}^{+} b_{\mathbf{q}} + \text{H.c.} \tag{2.2}$$

In a local description (Wannier representation) the corresponding operators are

$$c_{i\sigma}^{+} = L^{-1/2} \sum_{\mathbf{k}} a_{\mathbf{k}\sigma}^{+} e^{-i\mathbf{k}\cdot\mathbf{R}_i}; \quad d_l = L^{-1/2} \sum_{\mathbf{q}} b_{\mathbf{q}} e^{i\mathbf{q}\cdot\mathbf{R}_l}, \tag{2.3}$$

where $\mathbf{R}_i$ is a vector of the CuO_2 lattice and the interaction (2.2) becomes

$$H' = a\gamma \sum_{i,j,l} \tilde{\varphi}(\mathbf{R}_i - \mathbf{R}_j)\, \delta_{\mathbf{R}_l, (\mathbf{R}_i + \mathbf{R}_j)/2} c_{i\uparrow}^{+} c_{j\downarrow}^{+} d_l + \text{H.c.} \tag{2.4}$$

From the δ function in this expression one sees that there is no (static) dipol moment between the centers of charge of the carrier pair and of the local boson. This fact also allows us to make an estimate of the coupling constant γ. Indeed, assuming point charges the Coulomb energy between the three bodies is $-7e^2/\varepsilon a$ where the dielectric constant ε corrects for the assumption of point charges. With $\varepsilon \sim 30$ and $a \sim 3.3$ Å this leads to $\gamma \sim 1$ eV. This estimate shows the dominance of a charge-transfer mechanism over a spin-coupling scenario.[16] More explicitly, the ratio between magnetic and electric energies is of the order $(2\mu_B/ea)^2 = (\lambda_c/2\pi a)^2 \sim 10^{-6}$ where μ_B is the Bohr magneton and λ_c the Compton wavelength.

The assumption of local pairs now is expressed by the following form of the pair wave function:

$$\tilde{\varphi}(\mathbf{R}_i - \mathbf{R}_j) = \frac{\sqrt{L}}{2a} \sum_{\boldsymbol{\delta}} \sum_{\mu = 0,1,2,\ldots} \alpha_\mu e^{i\mu\vartheta(\boldsymbol{\delta})} \delta_{\mathbf{R}_i - \mathbf{R}_j, \boldsymbol{\delta}} \tag{2.5}$$

where $\boldsymbol{\delta}$ is one of the four nearest-neighbor vectors $(\pm a, 0)$, $(0, \pm a)$ in the (x,y) plane, $\vartheta(\boldsymbol{\delta})$ is the angle between $\boldsymbol{\delta}$ and the x axis (a multiple of $\pi/2$) and μ represents the angular momentum perpendicular to the CuO_2 plane. Singlet pairing requires invariance of the CuO_2 lattice with respect to reflections $\boldsymbol{\delta} \to -\boldsymbol{\delta}$. But this excludes odd values $\mu = 1,3,\ldots$ so that, because of the periodicity modulo 4 in μ, only $\mu = 0$ and 2, i.e., s- and d-symmetric pairings, remain in Eq. (2.5). While s-wave pairs belong to a trivial representation of the rotation symmetry of the CuO_2 lattice (rotations by multiples of $\pi/2$), the representation of the d-wave pairs ''breaks'' this symmetry.

The interpretation of the explicitly written term of Eq. (2.4) is the nominal charge transfer[10,11]

$$\mathrm{Cu}_{i\uparrow}^{2+} \mathrm{O}_l^{0} \mathrm{Cu}_{j\downarrow}^{2+} \to \mathrm{Cu}_{i\uparrow\downarrow}^{3+} \mathrm{O}_l^{2-} \mathrm{Cu}_{j\uparrow\downarrow}^{3+}. \tag{2.6}$$

Physically, the created holes, instead of being localized on the coppers will rather be distributed on the four surrounding oxygens,[17] while the doubly charged boson describing the neutralizing polarization charge may even be distributed outside of the CuO_2 planes (e.g., on the apex oxygens) but centered at O_l. Equation (2.6) corresponds to a $d^9 \to d^8$ charge transfer. On the other hand, the evidence from optical experiments seems to be $d^9 \to d^{10}$.[5,6] An interpretation of such a transfer, analogous to Eq. (2.6), would be

$$\mathrm{Cu}_{i\uparrow}^{2+} \mathrm{O}_l^{2-} \mathrm{Cu}_{j\downarrow}^{2+} \to \mathrm{Cu}_i^{+} \mathrm{O}_l^{0} \mathrm{Cu}_j^{+}, \tag{2.7}$$

which is described by the H.c. term in Eq. (2.4). However, this transfer is unphysical since the left-hand side of Eq. (2.7) corresponds to the equilibrium configuration of the undoped cuprates, and hence Eq. (2.7) cannot describe the optical measurements. Therefore, the interpretation of the optical experiments in the framework of our model seems to require farther-than-nearest-neighbor pairings, as alluded to in the Introduction.

Fourier transforming Eq. (2.5) according to $\varphi_{\mathbf{k}} = (a/\sqrt{L}) \Sigma_i \tilde{\varphi}(\mathbf{R}_i) e^{-i\mathbf{k}\cdot\mathbf{R}_i}$ yields

$$\varphi_{\mathbf{k}} = \alpha_s \varphi_{s\mathbf{k}} + \alpha_d \varphi_{d\mathbf{k}}, \tag{2.8}$$

where

$$\left.\begin{matrix} \varphi_{s\mathbf{k}} \\ \varphi_{d\mathbf{k}} \end{matrix}\right\} = \frac{1}{L} \left[\cos(k_x a) \pm \cos(k_y a)\right] \tag{2.9}$$

are real functions satisfying the orthonormality relations

$$\sum_{\mathbf{k}} \varphi_{\mu\mathbf{k}} \varphi_{\mu'\mathbf{k}} = \delta_{\mu\mu'}, \tag{2.10}$$

which are a consequence of the identity $\Sigma_{\mathbf{k}} \cos(k_x a)\cos(k_y a) = 0$. The orthogonality expresses the fact that $\varphi_{s\mathbf{k}}$ and $\varphi_{d\mathbf{k}}$ are different irreducible representations of the square symmetry of the Brillouin zone (rotations by $\pi/2$ and reflections). Normalization, $\Sigma_{\mathbf{k}} |\varphi_{\mathbf{k}}|^2 = 1$, then implies

$$|\alpha_s|^2 + |\alpha_d|^2 = 1. \tag{2.11}$$

Note that in the absence of a pair current the phase of $\tilde{\varphi}(\mathbf{r})$ is not a function of $\mathbf{r}$ and hence may be chosen to be zero so that $\tilde{\varphi}(\mathbf{r})$ is real. From the symmetry $\varphi_{\mu-\mathbf{k}} = \varphi_{\mu\mathbf{k}}$ it then follows that $\varphi_{\mathbf{k}}$ and hence α_s and α_d may also be chosen to be real. However, since tunneling experiments are of crucial importance for the determination of the pairing symmetry,[4] this choice is only good in equilibrium.

Limitation of the kinetic energy

$$H_0 = \sum_{\mathbf{k}\sigma} \varepsilon_{\mathbf{k}} a_{\mathbf{k}\sigma}^{+} a_{\mathbf{k}\sigma} = \sum_{ij\sigma} t(\mathbf{R}_i - \mathbf{R}_j) c_{i\sigma}^{+} c_{j\sigma} \tag{2.12}$$

to nearest-neighbor hoppings, $t(\mathbf{R}_i) = -t\,\delta_{\mathbf{R}_i, \boldsymbol{\delta}}$, leads to the usual band structure

$$\varepsilon_{\mathbf{k}} = -2t[\cos(k_x a) + \cos(k_y a)]. \tag{2.13}$$

We assume that due to a repulsive Hubbard term the band is defined by $-4t < \varepsilon_{\mathbf{k}} < 0$ and that the Fermi energy is close to half-filling, i.e., $0 < -\varepsilon_F \ll t$. This means that the densities of states (DOS's)

$$N^{(p)}(\lambda) = \frac{1}{La^2} \sum_{\mathbf{k}} \left(L\, \frac{\varphi_{s\mathbf{k}}^2 - \varphi_{d\mathbf{k}}^2}{4} \right)^p \delta(L\varphi_{s\mathbf{k}} - \lambda); \quad p = 0,1,2 \tag{2.14}$$

vanish by definition outside the interval $0 < \lambda < 2$.

III. WEAK-COUPLING THEORY

In weak-coupling approximation one proceeds by eliminating the boson field from the interaction (2.2) in the usual way[8] which results in an effective carrier attraction (in Ref. 11 a factor of 1/2 is missing)

$$H_{\rm attr}=\frac{|\gamma|^2}{2}\sum_{\mathbf{k}\mathbf{k}'\mathbf{q}}\varphi_{\mathbf{k}-\mathbf{q}/2}\varphi^*_{\mathbf{k}'-\mathbf{q}/2}\left\{\frac{1}{\varepsilon_{\mathbf{k}-\mathbf{q}}+\varepsilon_{\mathbf{k}}-\Omega_{\mathbf{q}}-i\epsilon}+\frac{1}{\varepsilon_{\mathbf{k}'-\mathbf{q}}+\varepsilon_{\mathbf{k}'}-\Omega_{\mathbf{q}}+i\epsilon}\right\}a^+_{\mathbf{k}\uparrow}a^+_{-\mathbf{k}+\mathbf{q}\downarrow}a_{-\mathbf{k}'+\mathbf{q}\downarrow}a_{\mathbf{k}'\uparrow}, \tag{3.1}$$

where $\Omega_{\mathbf{q}}$ is the frequency of the local boson. We now identify this mode with a feature seen in optical experiments at $\Omega_0\sim 1.6$ eV having a width of $\Delta\Omega\sim 0.1$ eV.[6] Since Ω_0 is an order of magnitude larger than both, $\Delta\Omega$ and the band energies $\varepsilon_{\mathbf{k}}$ involved, Eq. (3.1) may be expressed in terms of the pair operator Eq. (2.1) as

$$H_{\rm attr}=-g\sum_{\mathbf{q}}B^+_{\mathbf{q}}B_{\mathbf{q}}, \tag{3.2}$$

where $g=|\gamma|^2/\Omega_0\sim 0.6$ eV. With this expression one immediately obtains the gap equation[8]

$$\Delta_{\mathbf{k}}=\frac{g}{2}\sum_{\mathbf{k}'}(\varphi_{\mathbf{k}}\varphi^*_{\mathbf{k}'}+\varphi^*_{\mathbf{k}}\varphi_{\mathbf{k}'})\frac{\Delta_{\mathbf{k}'}}{2E_{\mathbf{k}'}}\tanh\frac{\beta E_{\mathbf{k}'}}{2}\times\Theta(\Delta\Omega-|\xi_{\mathbf{k}'}|), \tag{3.3}$$

where β^{-1} is the temperature in energy units, $E^2_{\mathbf{k}}=\xi^2_{\mathbf{k}}+\Delta^2_{\mathbf{k}}$ and $\xi_{\mathbf{k}}=\varepsilon_{\mathbf{k}}-\varepsilon_F$, and where an *ad hoc* cutoff by the boson line width $\Delta\Omega$ has been introduced in analogy to the Debye frequency used in BCS theory. It turns out that this cutoff is a necessary approximation and also a device to control the van Hove singularities at $a\mathbf{k}=(\pm\pi,0)$, $(0,\pm\pi)$ by choosing $\Delta\Omega<|\varepsilon_F|$. It may be justified by assuming the off-shell energy $\Omega_{\mathbf{q}}-\varepsilon_{\mathbf{k}-\mathbf{q}}-\varepsilon_{\mathbf{k}}$ in Eq. (3.1) to be approximately independent of $\mathbf{q}$. Indeed, by subtracting its value for $\mathbf{q}=0$ one obtains $\varepsilon_{\mathbf{k}-\mathbf{q}}-\varepsilon_{\mathbf{k}}\sim\Omega_{\mathbf{q}}-\Omega_0$ or, with $\varepsilon_{\mathbf{k}}\sim\varepsilon_F$, $|\xi_{\mathbf{k}}|<\Delta\Omega$. In equilibrium we may take $\varphi_{\mathbf{k}}$ to be real so that, with $\Delta_{\mathbf{k}}=\varphi_{\mathbf{k}}\Delta$, Eq. (3.3) becomes

$$1=g\sum_{\mathbf{k}}\varphi^2_{\mathbf{k}}\frac{\tanh(\beta E_{\mathbf{k}}/2)}{2E_{\mathbf{k}}}\Theta(\Delta\Omega-|\xi_{\mathbf{k}}|) \tag{3.4}$$

and the transition temperature T_c is obtained in the limit $\Delta\to 0$.

According to Eq. (2.8), Eq. (3.4) contains both, s- and d-wave pairing contributions[12,18] and, for $\alpha_s=0$ and without cutoff, reduces to the gap equation of Ref. 19. Now it follows from square symmetry that, for any function F, $\Sigma_{\mathbf{k}}\varphi_{s\mathbf{k}}\varphi_{d\mathbf{k}}F(\xi_{\mathbf{k}})=0$. Making use of Eqs. (2.8) and (2.11) one then sees that the linearized ($\Delta\to 0$) gap equation (3.4) decomposes (as it should for different irreducible representations) into the sum of the gap equations

$$1=g\sum_{\mathbf{k}}\varphi^2_{\mu\mathbf{k}}\frac{\tanh(\beta E_{\mu\mathbf{k}}/2)}{2E_{\mu\mathbf{k}}}\Theta(\Delta\Omega-|\xi_{\mathbf{k}}|);\quad \mu=s,d, \tag{3.5}$$

linearized according to $\overline{\Delta}_{\mu}\to 0$, where $E^2_{\mu\mathbf{k}}=\xi^2_{\mathbf{k}}+\Delta^2_{\mu\mathbf{k}}$ and $\Delta_{\mu\mathbf{k}}=\varphi_{\mu\mathbf{k}}\overline{\Delta}_{\mu}$. Equations (3.5) determine the s- and d-wave order parameters $\Delta_{s\mathbf{k}}$ and $\Delta_{d\mathbf{k}}$ and, in the limit $\overline{\Delta}_{\mu}\to 0$, the corresponding transition temperatures T_{cs} and T_{cd}. Using the DOS's (2.14) in Eqs. (3.5) these transition temperatures are determined, respectively, by

$$\frac{4t}{a^2g}=\int_{-\Delta\omega}^{+\Delta\omega}d\omega\,N^{(0)}\left(\frac{\omega+\omega_F}{\beta t}\right)\left(\frac{\omega+\omega_F}{\beta t}\right)^2\frac{\tanh\omega}{\omega} \tag{3.6}$$

and

$$\frac{4t}{a^2g}=\int_{-\Delta\omega}^{+\Delta\omega}d\omega\left\{N^{(0)}\left(\frac{\omega+\omega_F}{\beta t}\right)\left(\frac{\omega+\omega_F}{\beta t}\right)^2-4N^{(1)}\left(\frac{\omega+\omega_F}{\beta t}\right)\right\}\frac{\tanh\omega}{\omega}, \tag{3.7}$$

where $\omega_F\equiv(\beta/2)|\varepsilon_F|$ and $\Delta\omega\equiv(\beta/2)\Delta\Omega$.

Although the DOS's $N^{(p)}(\lambda)$ are given by elliptic integrals[19] an approximate evaluation is possible in the limit $\lambda\ll 1$ by dividing the energy contours into sections dominated by the van Hove singularities and straight sections dominated by nesting. Inserting the results Eqs. (A4) and (A5) derived in the Appendix into Eqs. (3.6) and (3.7) one finds, neglecting ω in the arguments $\omega+\omega_F$,

$$T_{cs}\simeq 1.13\Delta\Omega\,\exp\left\{-\frac{8\pi^2t^3}{g|\varepsilon_F|^2[\ln(\pi^2t/4|\varepsilon_F|)+\pi/2]}\right\} \tag{3.8}$$

and

$$T_{cd}\simeq 1.13\Delta\Omega\times\exp\left\{-\frac{\pi^2t}{2g[\ln(\pi^2t/4|\varepsilon_F|)-(\pi/4)^2+\pi^3/96]}\right\}. \tag{3.9}$$

With $t\sim 0.5$ eV, $|\varepsilon_F|\sim\Delta\Omega\sim 0.1$ eV, $g\sim 0.6$ eV one finds $T_{cs}\sim 10^{-172}$ K and $T_{cd}\sim 205$ K. In the same approximation the solution of Eq. (3.4) in the limit $\Delta\to 0$ may be written

$$T_c\simeq 1310\ \text{K}\times\exp\left\{-\frac{1}{0.00248\alpha_s^2+0.540\alpha_d^2}\right\}, \tag{3.10}$$

which shows that, for any real values of α_s and α_d satisfying Eq. (2.11),

$$T_{cs}\leqslant T_c\leqslant T_{cd}. \tag{3.11}$$

More generally, there may be crossings between the three transition temperatures as functions of the chemical potential ε_F (see the third Ref. 12). The contributions to Eqs. (3.4) from the region $\Delta\Omega<-\xi_{\mathbf{k}}<4t-|\varepsilon_F|$ outside the cutoff may be estimated by using Eqs. (A8). One finds that T_{cs} is raised by 71 orders while T_{cd} increases by a factor of ~ 16 to a totally unphysical value. This shows that, at least in the above approximation, the cutoff is necessary.

It is interesting that the physically relevant transition temperature T_{cd} is a simple function of $|\varepsilon_F|$; from Eq. (3.9) one finds

$$\frac{dT_{cd}}{d|\varepsilon_F|}\simeq-\frac{T_{cd}}{|\varepsilon_F|}\frac{\pi^2t}{2g}\left[\ln\left(\frac{2t\lambda_d}{|\varepsilon_F|}\right)\right]^{-2}. \tag{3.12}$$

On the other hand, using Eq. (A4), the hole density is given by

$$n_h = 2t\int_0^{|\varepsilon_F|/2t} d\lambda\, N^{(0)}(\lambda) \simeq \frac{|\varepsilon_F|}{\pi^2 a^2}\left\{1+\frac{\pi}{2}+\ln\left(\frac{\pi^2 t}{4|\varepsilon_F|}\right)\right\}. \tag{3.13}$$

Hence

$$\frac{dn_h}{d|\varepsilon_F|} \simeq \frac{1}{\pi^2 a^2}\left\{\frac{\pi}{2}+\ln\left(\frac{\pi^2 t}{4|\varepsilon_F|}\right)\right\} \tag{3.14}$$

and, combining with Eq. (3.12), one concludes that $dT_{cd}/dn_h<0$. Thus our approximation seems to be appropriate in the region where doping leads to a decrease of T_c and at the same time where the Uemura line $T_c(n_s)$ bends downward.[20]

IV. THE GINZBURG-LANDAU FREE ENERGY

Leaving the discussion of the gap equation to Sec. V where a strong-coupling form will be given, we first wish to derive a Ginzburg-Landau functional for the combined s- and d-wave pairing described above. But rather than using the Gor'kov method[18,8] we propose to proceed by the calculation of an effective action, eliminating the carrier operators via a Hubbard-Stratonovich transformation. It turns out, in fact, that the form (3.2) of the effective attraction is the most general form of an interaction that may be subject to such a transformation.[14] Leaving out the details[14] we may express the transformed partition function $Z=\mathrm{Tr}\exp[-\beta(H_0+H')]$ as functional integral over the auxiliary variables which here are the time-dependent order parameters $\Delta_{s\mathbf{q}}(\tau)$ and $\Delta_{d\mathbf{q}}(\tau)$ and their complex conjugates,

$$\frac{Z}{Z_0} = \int\int D^2\Delta_s D^2\Delta_d \times \exp\left\{-\int_0^\beta d\tau \sum_{\mu\mathbf{q}} \frac{1}{g}|\Delta_{\mu\mathbf{q}}(\tau)|^2\right\} W[\Delta_s,\Delta_d], \tag{4.1}$$

where $Z_0=\mathrm{Tr}\exp(-\beta H_0)$, τ is the imaginary time and

$$W \equiv \left\langle T\exp\left\{\int_0^\beta d\tau \sum_{\mu\mathbf{k}\mathbf{q}} [\varphi_{\mu\mathbf{k}-\mathbf{q}/2}\Delta_{\mu\mathbf{q}}(\tau) a^+_{\mathbf{k}\uparrow} a^+_{\mathbf{q}-\mathbf{k}\downarrow} + \mathrm{H.c.}](-i\tau)\right\}\right\rangle. \tag{4.2}$$

In the last expression $-i\tau$ is the argument of the interaction representation $A(-i\tau)=e^{H_0\tau}Ae^{-H_0\tau}$.

Defining the effective action S by the relation $Z/Z_0=\int\int D^2\Delta_s D^2\Delta_d \exp(-S)$ one has

$$S[\Delta_s,\Delta_d] = \int_0^\beta d\tau \sum_{\mu\mathbf{q}} \frac{1}{g}|\Delta_{\mu\mathbf{q}}(\tau)|^2 - \ln W, \tag{4.3}$$

where $\ln W$ now is given by the same expression (4.2) but restricted to connected diagrams. It is easy to see that, due to the form (2.1) of the operator $B^+_{\mathbf{q}}$ the only connected diagrams are closed loops with an even number of alternating two-leg vertices[14] labeled by $v\equiv\Sigma_\mu \varphi_{\mu\mathbf{k}-\mathbf{q}/2}\tilde{\Delta}_\mu(q)$ and v^* where $q\equiv(\mathbf{q},iq_0)$ and

$$\tilde{\Delta}_\mu(\mathbf{q},iq_0) = \beta^{-1}\int_0^\beta d\tau\, \Delta_{\mu\mathbf{q}}(\tau)e^{iq_0\tau}. \tag{4.4}$$

To fourth order the result may be cast into the following form:

$$S = \sum_q \sum_{\mu\mu'}\left\{\frac{\beta}{g}\delta_{\mu\mu'} - A_{\mu\mu'}(q)\right\}\tilde{\Delta}_\mu(q)\tilde{\Delta}^*_{\mu'}(q) + \sum_{q,q',q''}\sum_{\mu\mu'\nu\nu'} B_{\mu\mu'\nu\nu'}(q,q',q'')\tilde{\Delta}_\mu(q)\tilde{\Delta}^*_{\mu'}(q')\tilde{\Delta}_\nu(q'') \times\tilde{\Delta}^*_{\nu'}(q-q'+q''). \tag{4.5}$$

In terms of the unperturbed carrier propagator

$$G(k) = \int_0^\beta d\tau\langle T[a_{\mathbf{k}}(-i\tau)a^+_{\mathbf{k}}(0)]\rangle e^{ik_0\tau} = \frac{1}{ik_0-\xi_{\mathbf{k}}}, \tag{4.6}$$

where $k\equiv(\mathbf{k},ik_0)$ one finds

$$A_{\mu\mu'}(q) = \sum_k \varphi_{\mu\mathbf{k}-\mathbf{q}/2}\varphi_{\mu'\mathbf{k}-\mathbf{q}/2}G(k)G(q-k) \tag{4.7}$$

and

$$B_{\mu\mu'\nu\nu'}(q,q',q'') = 2\sum_k \varphi_{\mu\mathbf{k}-\mathbf{q}/2}\varphi_{\mu'\mathbf{k}-\mathbf{q}'/2}\varphi_{\nu\mathbf{k}-\mathbf{q}'+\mathbf{q}''/2}\varphi_{\nu'\mathbf{k}-(\mathbf{q}+\mathbf{q}'-\mathbf{q}'')/2} \times G(k)G(k-q'+q'')G(q-k)G(q'-k). \tag{4.8}$$

Retaining in $A_{\mu\mu'}$ terms up to second order in $\mathbf{q}$ and up to first order in q_0 and dropping any q dependence in $B_{\mu\mu'\nu\nu'}$ the effective action Eq. (4.5) becomes the time-dependent Ginzburg-Landau functional.[13] (The results of Refs. 13 are calculated for a single order parameter and with $\varphi_{\mathbf{k}}=1$.)

Summing first over k_0 and then shifting the summation variable from $\mathbf{k}$ to $\mathbf{k}+\mathbf{q}/2$, Eq. (4.7) takes the form

$$A_{\mu\mu'}(q) = \beta\sum_{\mathbf{k}} \varphi_{\mu\mathbf{k}}\varphi_{\mu'\mathbf{k}}\frac{1-f_0(\xi_{\mathbf{k}+\mathbf{q}/2})-f_0(\xi_{\mathbf{k}-\mathbf{q}/2})}{\xi_{\mathbf{k}+\mathbf{q}/2}+\xi_{\mathbf{k}-\mathbf{q}/2}-iq_0}, \tag{4.9}$$

which is manifestly even in $\mathbf{q}$. In this paper we are interested only in the equilibrium version of the effective action which is β times the Ginzburg-Landau free energy. For time-independent order parameters $\Delta_{\mu\mathbf{k}}$ it follows from Eq. (4.4) that $\tilde{\Delta}_\mu(\mathbf{q},iq_0)=\Delta_{\mu\mathbf{q}}\delta_{q_0,0}$ so that we may set $q_0=0$ in Eq. (4.9). To second order in $\mathbf{q}$ one then finds

$$A_{\mu\mu'}(\mathbf{q},0) = \beta\Gamma_\mu(T)\delta_{\mu\mu'} + \mathbf{q}\cdot\mathbf{K}_{\mu\mu'}\mathbf{q}. \tag{4.10}$$

Here

$$\Gamma_\mu(T) = \frac{\beta}{4}\sum_{\mathbf{k}} \varphi^2_{\mu\mathbf{k}}\frac{\tanh\omega_{\mathbf{k}}}{\omega_{\mathbf{k}}} \tag{4.11}$$

and the $\mathbf{K}_{\mu\mu'}$ are 2×2 matrices given by

$$\mathbf{q}\cdot\mathbf{K}_{\mu\mu'}\mathbf{q}=\frac{\beta^2}{32}\sum_{\mathbf{k}}\varphi_{\mu\mathbf{k}}\varphi_{\mu'\mathbf{k}}\frac{1}{\omega_{\mathbf{k}}}\left\{[(\mathbf{q}\cdot\partial_{\mathbf{k}})^2\omega_{\mathbf{k}}]\left[\frac{1}{\cosh^2\omega_{\mathbf{k}}}-\frac{\tanh\omega_{\mathbf{k}}}{\omega_{\mathbf{k}}}\right]-2[(\mathbf{q}\cdot\partial_{\mathbf{k}})\omega_{\mathbf{k}}]^2\frac{\tanh\omega_{\mathbf{k}}}{\cosh^2\omega_{\mathbf{k}}}\right\},\quad(4.12)$$

where $\omega_{\mathbf{k}}\equiv\beta\xi_{\mathbf{k}}/2$. In the case where all $q=0$ expression (4.8) becomes after summation over k_0

$$B_{\mu\mu'\nu\nu'}(0)=\frac{\beta^4}{16}\sum_{\mathbf{k}}\varphi_{\mu\mathbf{k}}\varphi_{\mu'\mathbf{k}}\varphi_{\nu\mathbf{k}}\varphi_{\nu'\mathbf{k}}\times\frac{1}{\omega_{\mathbf{k}}^2}\left[\frac{\tanh\,\omega_{\mathbf{k}}}{\omega_{\mathbf{k}}}-\frac{1}{\cosh^2\,\omega_{\mathbf{k}}}\right].\quad(4.13)$$

For further evaluation of Eqs. (4.12) and (4.13) we first symmetrize with respect to the square symmetry of the Brillouin zone. Indicating this operation by brackets and introducing the abbreviations $c_i(\mathbf{k})\equiv\cos(k_ia)$ and $s_i(\mathbf{k})\equiv\sin(k_ia)$ $(i=x,y)$ so that $(\mathbf{q}\cdot\partial_{\mathbf{k}})^2\omega_{\mathbf{k}}=\beta ta^2\Sigma_i q_i^2c_i$ and $[(\mathbf{q}\cdot\partial_{\mathbf{k}})\omega_{\mathbf{k}}]^2=(\beta ta\Sigma_i q_is_i)^2$ one finds

$$\left\langle\varphi_{\mu\mathbf{k}}^2\sum_i q_i^2c_i\right\rangle=\frac{1}{2}\varphi_{\mu\mathbf{k}}^2(c_x+c_y)(q_x^2+q_y^2),\quad(4.14)$$

$$\left\langle\varphi_{s\mathbf{k}}\varphi_{d\mathbf{k}}\sum_i q_i^2c_i\right\rangle=\frac{1}{2}\varphi_{s\mathbf{k}}\varphi_{d\mathbf{k}}(c_x-c_y)(q_x^2-q_y^2),$$

$$\left\langle\varphi_{\mu\mathbf{k}}^2\left(\sum_i q_is_i\right)^2\right\rangle=\frac{1}{2}\varphi_{\mu\mathbf{k}}^2(s_x^2+s_y^2)(q_x^2+q_y^2),\quad(4.15)$$

$$\left\langle\varphi_{s\mathbf{k}}\varphi_{d\mathbf{k}}\left(\sum_i q_is_i\right)^2\right\rangle=\frac{1}{2}\varphi_{s\mathbf{k}}\varphi_{d\mathbf{k}}(s_x^2-s_y^2)(q_x^2-q_y^2),$$

and

$$\langle(\varphi_{\mu\mathbf{k}}\varphi_{\mu'\mathbf{k}})^2\rangle=(\varphi_{\mu\mathbf{k}}\varphi_{\mu'\mathbf{k}})^2;\quad\langle\varphi_{\mu\mathbf{k}}^3\varphi_{\nu\mathbf{k}}\rangle=0;\quad\mu\neq\nu.\quad(4.16)$$

Using these expressions in Eqs. (4.10)–(4.13) the action (4.5) may be written in the form of Eq. (1) of Sigrist *et al.* (second Ref. 4),

$$F[\Delta_s,\Delta_d]=\sum_{\mathbf{q}}\left[\sum_{\mu=s,d}\{\tilde{a}_\mu(T)|\Delta_{\mu\mathbf{q}}|^2+b_\mu|\Delta_{\mu\mathbf{q}}|^4+K_\mu|\mathbf{q}\Delta_{\mu\mathbf{q}}|^2\}+\gamma_1|\Delta_{s\mathbf{q}}|^2|\Delta_{d\mathbf{q}}|^2+\frac{\gamma_2}{2}(\Delta_{s\mathbf{q}}^{*2}\Delta_{d\mathbf{q}}^2+\text{c.c.})+\frac{\tilde{K}}{2}\{(q_x\Delta_{s\mathbf{q}})^*q_x\Delta_{d\mathbf{q}}-(q_y\Delta_{s\mathbf{q}})^*q_y\Delta_{d\mathbf{q}}+\text{c.c.}\}\right].\quad(4.17)$$

Here $\tilde{a}_\mu(T)=1/g-\Gamma_\mu(T)$, $b_\mu=\beta^{-1}B_{\mu\mu\mu\mu}(0)$, $\gamma_1=2\beta^{-1}B_{ssdd}(0)$, $\gamma_2=2\beta^{-1}B_{sdsd}(0)$, $K_\mu=K_{\mu\mu}^{(+)}$, and $\tilde{K}=2K_{sd}^{(-)}$, where

$$K_{\mu\mu'}^{(\pm)}\equiv-\frac{\beta^2ta^2}{64}\sum_{\mathbf{k}}\varphi_{\mu\mathbf{k}}\varphi_{\mu'\mathbf{k}}\frac{1}{\omega_{\mathbf{k}}}\left\{\left[\frac{1}{\cosh^2\omega_{\mathbf{k}}}-\frac{\tanh\omega_{\mathbf{k}}}{\omega_{\mathbf{k}}}\right]\times(c_x\pm c_y)-\beta t\frac{\tanh\omega_{\mathbf{k}}}{\cosh^2\omega_{\mathbf{k}}}(s_x^2\pm s_y^2)\right\}.\quad(4.18)$$

Note that here $\gamma_1=\gamma_2$ and that our result is essentially the same as Eq. (16) of Ref. 18. $\tilde{a}_\mu(T)=0$ $(\mu=s,d)$ are the gap equations (3.5) (without cutoff) in the limit $\Delta_\mu\to0$ determining the transition temperatures $T_{c\mu}$.

V. STRONG-COUPLING THEORY

For the calculation of the self-energies we follow Ref. 11 by first writing the carrier-boson interaction Eq. (2.2) in the Nambu representation,

$$H'=\sum_{\mathbf{kq}}\varphi_{\mathbf{k}-\mathbf{q}/2}\Psi_{\mathbf{k}}^+\{\gamma P_+b_{\mathbf{q}}+\gamma^*P_-b_{-\mathbf{q}}^+\}\Psi_{\mathbf{k}-\mathbf{q}},\quad(5.1)$$

where

$$\Psi_{\mathbf{k}}=\begin{Bmatrix}a_{\mathbf{k}\uparrow}\\a_{-\mathbf{k}\downarrow}^+\end{Bmatrix}\quad(5.2)$$

and $P_\pm\equiv(\tau_1\pm i\tau_2)/2$, $Q_\pm\equiv(1\pm\tau_3)/2$, τ_i $(i=1,2,3)$ being the Pauli matrices. The carrier self-energy $\Sigma(k)$ then consists of a normal Hartree-type contribution

$$\Sigma_H(k)=\frac{|\gamma|^2}{\beta}\sum_q D(q)\{\varphi_{\mathbf{k}-\mathbf{q}/2}^2G(q-k)Q_+-\varphi_{\mathbf{k}+\mathbf{q}/2}^2G(k+q)Q_-\}\quad(5.3)$$

and an anomalous exchange-type part

$$\Sigma_E(k)=\frac{|\gamma|^2}{\beta}D(0)\varphi_{\mathbf{k}}\sum_{k'}\varphi_{\mathbf{k}'}\{F(k')P_++F^*(k')P_-\},\quad(5.4)$$

where $D(q)$ is the boson propagator and $F(k)$ the Gor'kov function as defined by the renormalized carrier Green's function in the Nambu representation,

$$\mathbf{G}(k)=F(k)P_++F^*(k)P_-+G_\uparrow(k)Q_++G_\downarrow(k)Q_-.\quad(5.5)$$

Writing its reciprocal as

$$\begin{aligned}\mathbf{G}^{-1}(k)&=\mathbf{G}_0^{-1}(k)-\Sigma(k)\\&=\Delta_+(k)P_+ +\Delta_-(k)P_- +Z_+(k)Q_+\\&\quad +Z_-(k)Q_-,\end{aligned}\tag{5.6}$$

where $\mathbf{G}_0(k)=G(k)Q_+ -G(-k)Q_-$ and inserting expressions (5.3) and (5.4) one obtains the four equations[11]

$$\Delta_\pm(k)=-\frac{|\gamma|^2}{\beta}D(0)\varphi_{\mathbf{k}}\sum_q \varphi_{\mathbf{q}}\frac{\Delta_\pm(q)}{K(q)}\tag{5.7}$$

and

$$Z_\pm=ik_0\mp\xi_{\mathbf{k}}\pm\frac{|\gamma|^2}{\beta}\sum_q D(q)\varphi^2_{\mathbf{k}\mp\mathbf{q}/2}\frac{Z_\mp(q\mp k)}{K(q\mp k)},\tag{5.8}$$

where $K\equiv\Delta_+\Delta_- -Z_+Z_-$.

The solution of Eqs. (5.7) has the same form as in the weak-coupling approximation, $\Delta_\pm(k)=\varphi_{\mathbf{k}}\Delta\equiv\Delta_{\mathbf{k}}$, but the gap equation now is

$$1=-\frac{|\gamma|^2}{\beta}D(0)\sum_q\frac{\varphi^2_{\mathbf{q}}}{K(q)}.\tag{5.9}$$

Inserting here the zeroth approximation $Z^{(0)}_\pm(k)=ik_0\mp\xi_{\mathbf{k}}$ and $D^{(0)}(q)=1/(iq_0-\Omega_{\mathbf{q}})$, Eq. (3.4) is recovered but without the cutoff. Such a cutoff is most probably implicit in a simultaneous solution of the nonlinear system of equations (5.7) and (5.8) which we do not attempt [it does not show in a first iteration of Eqs. (5.8)].

Even in the strong-coupling form Eq. (5.7) the order parameters are independent of k_0 and hence do not contain retardation effects. This could be interpreted as an indication that Migdal's theorem is not valid.[21] Indeed, retardation may be included by adding vertex corrections in the carrier self-energy. To lowest order this is achieved by combining the diagrams representing the self-energies Eqs. (5.3) and (5.4). There is, however, a different interpretation. Indeed, Eqs. (3.9) and (3.13) indicate that the relevant transition temperature T_{cd} is a unique function of the hole density n_h as described by the empirical Uemura relation.[20] But, as argued in the first Ref. 12, this seems to be incompatible with a frequency-dependent order parameter and hence with retardation effects.

Turning now to the properties of the local boson they are contained in the renormalized propagator $D(q)$. From the boson self-energy[11]

$$\Pi(q)=-\frac{|\gamma|^2}{\beta}\sum_k \varphi^2_{\mathbf{k}-\mathbf{q}/2}G_\uparrow(k)G_\downarrow(q-k)\tag{5.10}$$

one obtains, substituting the propagators with the help of Eqs. (5.5) and (5.6),

$$D^{-1}(q)=iq_0-\Omega_0+\frac{|\gamma|^2}{\beta}\sum_k \varphi^2_{\mathbf{k}-\mathbf{q}/2}\frac{Z_-(k)Z_+(q-k)}{K(k)K(q-k)}.\tag{5.11}$$

Inserting here the zeroth approximation $Z^{(0)}_\pm$ and setting $\Delta=0$ one finds, after performing first the k_0 sum and then shifting the summation variable from $\mathbf{k}$ to $\mathbf{k}+\mathbf{q}/2$, the first iteration

$$D^{(1)-1}(q)=iq_0-\Omega_0+|\gamma|^2\sum_{\mathbf{k}}\varphi^2_{\mathbf{k}}\frac{f_0(\xi_{\mathbf{k}+\mathbf{q}/2})-f_0(\xi_{\mathbf{k}-\mathbf{q}/2})}{\xi_{\mathbf{k}+\mathbf{q}/2}-\xi_{\mathbf{k}-\mathbf{q}/2}-iq_0}.\tag{5.12}$$

Here both, numerator and denominator are manifestly odd functions of $\mathbf{q}$ so that expansion to order $\mathbf{q}^2$ yields, after analytic continuation $iq_0\rightarrow\omega$ and setting $\omega=\Omega_0$ in the denominator,

$$D^{(1)-1}(\mathbf{q},\omega)=\omega-\Omega_0-\frac{|\gamma|^2}{4\beta\Omega_0^2}\sum_{\mathbf{k}}\varphi^2_{\mathbf{k}}\frac{[(\mathbf{q}\cdot\partial_{\mathbf{k}})\omega_{\mathbf{k}}]^2}{\cosh^2\omega_{\mathbf{k}}}.\tag{5.13}$$

Therefore, using Eqs. (4.15) to symmetrize with respect to square symmetry, the renormalized boson frequency is found from $D^{(1)-1}=0$ to be

$$\Omega^{\rm ren}_{\mathbf{q}}=\Omega_0+M\mathbf{q}^2+\widetilde{M}(q_x^2-q_y^2),\tag{5.14}$$

where $M=\Sigma_\mu M^{(+)}_{\mu\mu}/2$, $\widetilde{M}=M^{(-)}_{sd}$ and, with $g=|\gamma|^2/\Omega_0$,

$$M^{(\pm)}_{\mu\mu'}\equiv\frac{\beta g}{\Omega_0}(ta)^2\sum_{\mathbf{k}}\varphi_{\mu\mathbf{k}}\varphi_{\mu'\mathbf{k}}\frac{s_x^2\pm s_y^2}{\cosh^2\omega_{\mathbf{k}}}.\tag{5.15}$$

Inserting the DOS's (2.14) into Eq. (5.15) one finds

$$\begin{aligned}\frac{M}{a^2}=\frac{\beta g}{\Omega_0}(ta)^2\int_0^2\frac{d\lambda}{\cosh^2[\beta t(\lambda-\lambda_F)]}\{&2\lambda^2(2-\lambda^2)N^{(0)}(\lambda)\\&-8(1-\lambda^2)N^{(1)}(\lambda)-8N^{(2)}(\lambda)\}\end{aligned}\tag{5.16}$$

and

$$\begin{aligned}\frac{\widetilde{M}}{a^2}=\frac{\beta g}{\Omega_0}(ta)^2\int_0^2\frac{\lambda^2 d\lambda}{\cosh^2[\beta t(\lambda-\lambda_F)]}\{&-\lambda^2N^{(0)}(\lambda)\\&+4N^{(1)}(\lambda)\},\end{aligned}\tag{5.17}$$

where $\lambda_F\equiv|\varepsilon_F|/2t\ll1$. Since only the region around λ_F is important, application of Eqs. (A4)–(A6) leads to

$$\frac{M}{a^2}\simeq\frac{gt}{\Omega_0}\left(1+\frac{\pi}{6}\right)\simeq0.29\ \text{eV}\tag{5.18}$$

and

$$\frac{\widetilde{M}}{a^2}\simeq\frac{2g|\varepsilon_F|^2}{\pi\Omega_0 t}\left\{-\ln\left(\frac{\pi^2 t}{4|\varepsilon_F|}\right)+\frac{\pi^2}{4}\left(1-\frac{\pi}{6}\right)\right\}\simeq0.0034\ \text{eV}.\tag{5.19}$$

M may be used to estimate the boson mass m^* from $M=\hbar^2/2m^*$. One finds $m^*\sim1.5m_e$ where m_e is the free-electron mass. This means that the response of the neutralizing polarization charge is extremely fast. Finally one may estimate the extension q^* of this mode in k space from $Mq^{*2}=\Delta\Omega$; the result is $q^*\sim0.19(\pi/a)$.

VI. CONCLUSION

In this paper we have attempted to describe the complex physics of the superconducting cuprates by a simple but plausible model involving a charge-exchange rather than a spin-exchange mechanism. The arguments in favor of such a mechanism are multiple. First of all, electric energies were shown to be much larger than magnetic ones. Second, the cuprates are known to be highly polarizable; $SrTiO_3$ is almost ferroelectric. And third, recent optical experiments see a high-energy mode which seems to be involved in the pairing mechanism.[6]

The model is based on the idea that carrier pairs in the cuprates must be strongly localized which is supported by the short coherence lengths as well as by the proximity of a Mott transition. In the case of a nearest-neighbor singlet pair wave function, symmetry arguments uniquely lead to a superposition of specific *s* and *d* waves with antiferromagnetic correlation. The gap equations resulting from this construction yielded transition temperatures $T_{cs}\sim 0$ and $T_{cd}\sim 205$ K. However, this relatively reasonable value of T_{cd} was obtained only by introducing an *ad hoc* cutoff which clearly shows the shortcomings of the approximation. On the other hand, the strong-coupling formalism developed subsequently suggests that such a cutoff may be inherent in the derived nonlinear system of equations.

A rather gratifying result of the model is the expression obtained for the Ginzburg-Landau free energy which has the same general form as the phenomenological formula obtained from symmetry arguments alone.[4] Overall the results derived in this paper by simple, easily verifiable order-of-magnitude calculations can be considered as encouraging.

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APPENDIX

For $0<\lambda\ll 1$ the energy contours have almost square shape so that the DOS's, Eq. (2.14) may be written as four times the sum of the respective contributions from the corner (van Hove singularity) region at $a\mathbf{k}=(\pi,0)$ and from the straight (nesting) region at $a\mathbf{k}=(\pi/2,\pi/2)$,

$$N^{(p)}(\lambda)=N^{(p)}_{\mathrm{vH}}(\lambda)+N^{(p)}_{\mathrm{ne}}(\lambda);\quad p=0,1,2, \tag{A1}$$

where

$$N^{(p)}_{\mathrm{vH}}(\lambda)=\frac{2}{\pi^2a^2}\int\int_0^{\pi/4}du\ dv(-\cos u\ \cos v)^p \times\delta(\cos v-\cos u-\lambda) \tag{A2}$$

and

$$N^{(p)}_{\mathrm{ne}}(\lambda)=\frac{1}{\pi^2a^2}\int\int_{-\pi/4}^{\pi/4}du'dv'(\sin u'\ \sin v')^p \times\delta(\sin u'+\sin v'-\lambda). \tag{A3}$$

The approximation now consists of retaining the sin and cos functions only to first and second order in their arguments, respectively. A straightforward calculation then yields, to lowest order in λ,

$$N^{(0)}_{\mathrm{vH}}(\lambda)\simeq\frac{1}{\pi^2a^2}\ln\left(\frac{\pi^2}{8\lambda}\right);\quad N^{(0)}_{\mathrm{ne}}(\lambda)\simeq\frac{1}{2\pi a^2}, \tag{A4}$$

$$N^{(1)}_{\mathrm{vH}}(\lambda)\simeq-\frac{1}{\pi^2a^2}\ln\left(\frac{\pi^2}{8\lambda}\right)+\frac{1}{16a^2};\quad N^{(1)}_{\mathrm{ne}}(\lambda)\simeq-\frac{\pi}{96a^2}, \tag{A5}$$

and

$$N^{(2)}_{\mathrm{vH}}(\lambda)\simeq\frac{1}{\pi^2a^2}\ln\left(\frac{\pi^2}{8\lambda}\right)-\frac{1}{8a^2};\quad N^{(2)}_{\mathrm{ne}}(\lambda)\simeq 0. \tag{A6}$$

Note that, in Eq. (A4), $N^{(0)}(\lambda)>0$ for $\lambda<5.9$ so that the approximation is consistent.

For $0<\lambda'\equiv 2-\lambda\ll 1$ the energy contours are almost circular so that it is advantageous to write Eq. (2.14) in polar coordinates. Retaining the cos functions again only to second order in their arguments one has

$$N^{(p)}(2-\lambda')\simeq\frac{1}{\pi^2a^2}\int_0^{2\pi}d\varphi\int_0^{\pi}r\ dr\left(1-\frac{r^2}{2}\cos^2\varphi\right)^p \times\left(1-\frac{r^2}{2}\sin^2\varphi\right)^p\delta\left(\frac{r^2}{2}-\lambda'\right). \tag{A7}$$

To low order in λ' this yields

$$N^{(p)}(2-\lambda')\simeq\frac{2}{\pi a^2}(1-p\lambda');\quad p=0,1,2. \tag{A8}$$

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